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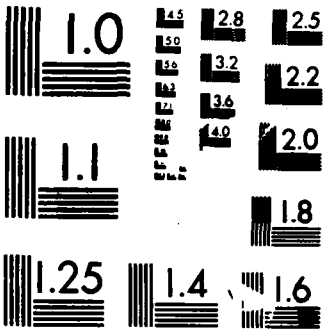

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This report summarizes research at the University of Mississippi in the area of Optical Sound Generation and Amplification during the period 1 January 1984 through 31 December 1986. Research has concentrated on sound propagation through a gas with a nonequilibrium distribution of internal states and the generation of sound following excitation of a fluid by a laser.

When a sound wave propagates through a gas which has an overpopulation of vibrationally excited states, the wave can increase in amplitude while propagating. In simple terms, this represents a reversal of the absorption typically associated with vibrational relaxation. Amplification of a propagating wave has been theoretically predicted and experimentally observed for a gas undergoing chemical reaction and following an electrical discharge through a non-reacting mixture.

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19. Abstract (continued)

Optoacoustic measurements have been completed in gaseous CO_2 and SF_6 and preliminary results are reported for several liquids. Following laser excitation of SF_6 at low pressure, the gas actually cooled. A theoretical model for this behavior consistent with known energy transfer mechanisms has been developed and shown to be consistent with experiment measurements.

PARGUM 87-1

OPTICAL SOUND GENERATION AND AMPLIFICATION

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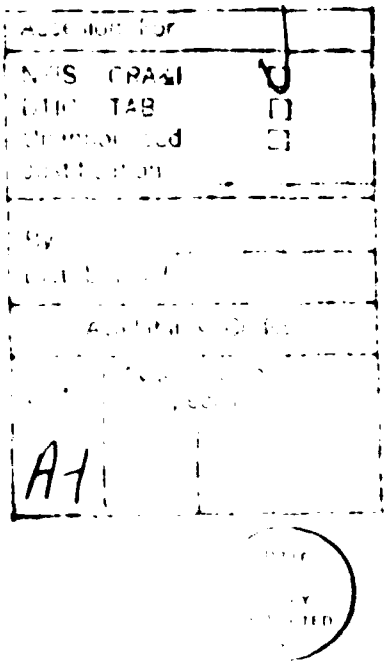

STUDY OF
OPTICAL SOUND GENERATION AND AMPLIFICATION

Background

The University of Mississippi study of Optical Sound Generation and Amplification began in 1981 as ONR contract N00014-81-K-0691. During the first 30 month period, this project involved Henry E. Bass, Lawrence Crum, and F. Douglas Shields as co-principal investigators. The first contract involved a shift in ongoing research in a number of areas of acoustics by the three principal investigators. As the previous efforts were carried to an orderly termination, a number of research reports and publications resulted.

Beginning in January of 1984, ONR contract N00014-84-C-0193 extended the Study of Optical Sound Generation and Amplification for an additional three year period. This contract expired 31 December 1986. This summary report presents a summary of research findings during this three year period. Where possible, the reader is referred to technical reports or publications for details. This report will deal with the general direction of research and most important results.

During this three year period five students were awarded Ph.D. degrees. One of these, Gary Hanson, is now working for the U.S. Army. Two, Anthony Atchley and Kerry Commander are working for the U.S. Navy. Two foreign students, Manaf Ali and Dadang Iskandor have returned to their home countries. In addition, a number of students have completed requirements for the M.S. degree. Although these students are not direct research results, their training represents a major product of this ongoing research program.

A brief review of publications resulting from the earlier contract will help to place more recent research in perspective. Due to the time lag between laboratory results and publication, the papers mentioned ap-

peared in print during this contract period. The description will be divided into sound amplification and sound generation.

Theoretical studies by Bass and Bauer[1] showed some years ago that a sound wave should be amplified upon passage through a gas with an over population of vibrationally excited states. They named this effect SACER (Sound Amplification from Controlled Excitation Reactions) due to similarity to the LASER.

Based upon the work of Gilbert[2], Bass chose to experimentally study amplification associated with chemical reactions. The system chosen for study was H_2/Cl_2 mixtures. The energy transfer rate constants required to predict amplification were measured independently and published in 1984.[3] Actual amplification was observed in 1983. These observations were published in 1985[4].

Basically, the phenomenon observed by Bass and Detsch (a former Ph.D. student now at NCSC) can be decided as follows. A UV lamp was used to photodissociate Cl_2. The resulting Cl atoms react very rapidly and exothermically to produce HCl. The rate of reaction depends upon the local pressure and temperature. Since the reaction is exothermic, an increase in reaction rate gives rise to a relative local increase in temperature, that is, local temperature and pressure variations are amplified.

Although the chemical SACER did exhibit gain, difficulties associated with controlling the chemical reaction and the complexity of the reaction scheme led us to abandon this effort in 1983 in favor of a system based solely on vibrational energy transfer in the gas. This approach entitled "Propagation of sound through gases with an over-population of vibrationally excited states" was being pursued simultaneously by Shields. His theoretical work published in 1984[5] indicated that amplification should be observed following an electrical discharge in N_2/He or N_2/H_2 mixtures.

This approach proved to be quite successful and is described in the following section.

While pursuing the problem of sound amplification in an excited gas, work was also underway investigating sound generation. The approach taken was the classic spectrophone. When a fluid is subjected to pulses of electromagnetic radiation of the proper wavelength, absorbed energy causes the gas to heat and expand resulting in a measurable acoustic impulse. Although the basic principles governing the spectrophone have been well established for many years[6], it was our goal to use this technique to study energy transfer at the molecular level. This required that other factors influencing the acoustic waveform were accurately included.

Although our long term goal was to examine dynamics in liquids, the first system studied under the previous contract was gaseous CO_2. The gas phase was chosen to give experimental control of the rate of energy transfer processes by varying the pressure. CO_2 was chosen because it weakly absorbed radiation from an existing CO_2 laser. Results of this effort were published in 1983[7]. This publication includes the first comprehensive theoretical treatment of the optoacoustic effect in a weakly absorbing (nearly transparent) fluid.

Optoacoustic studies in liquids typically involve a very short optical penetration depth. A gaseous analog to this situation is SF_6 absorbing 10.6 μ radiation from a CO_2 laser. A thorough study of this system was begun in 1983 and continued into the current contract period.

The effect of optical absorption is a laser beam which decreases in amplitude as it penetrates the fluid. Since the acoustic pressure generated is proportional to the incident optical intensity, the acoustic pressure is not a cylinder but, instead, it appears as an exponential decay rotated about the laser beam path. Since the acoustic pressure is greater near the optical entry point, in addition to the cylindrically spreading

wave (assumed for CO_2), there is an acoustic wave which propagates in the direction of the laser beam.

The results of the experimental and theoretical study of optoacoustic generation in SF_6 will be described in more detail later.

The foregoing was meant to provide the reader background for the current contract period and some rationale for the approaches taken. Accomplishments during the most recent three year period are reported in the following two sections; one devoted to Propagation of Sound Through a Gas with an Overpopulation of Vibrationally Excited States and one devoted to Optoacoustic Studies of Liquids. A third topic, Generation of Low Frequency Sound from Optical Pulses has received much less attention. A description of this task and results to date can be found in Ref. 8.

The Propagation of Sound Through a Gas with an Overpopulation of Vibrationally Excited States

The first step in this phase of the research program, as described in the previous section was to theoretically analyze a case that could be simulated in the laboratory. The theoretical work of Bauer and Bass[1] considered amplification while pumping molecules into the excited state. In order to simplify the analysis, we looked for a situation where a metastable vibrational state persisted long enough after the pumping mechanism was removed to allow the passage of a sound wave through the gas. Theoretical calculations of the magnitude of the expected sound amplifications for this situation in some specific gases were published in 1984[5].

These calculations showed that some very stringent conditions must be met if the effect was to be experimentally observed. For the magnitude of the gain to be appreciable, the v-t relaxation time could only be about one order of magnitude longer than the sound period. A number of experimental configurations have been tried to meet these conditions. Suffice it to say here that the effect has now been observed and measured in N_2/H_2, N_2/He and N_2/CH_4 mixtures.

Measurements of the absolute magnitude of the amplification as yet do not agree with calculated values, but the general dependence of the gain upon the vibrational relaxation time and vibrational temperature is as expected from theory.

Another discovery has been made in these experiments that was not expected. We have been able to measure the variation of the sound velocity in the gas as the vibrational energy decays into translation and from these measurements determine the variation of the translational temperature during the relaxation process. This in turn, allows a determination of the vibrational temperature as a function of time and, thus, the vibrational relaxation time and its temperature dependence. In the diatomic gases

currently under study (N_2 and CO) these numbers are very difficult to measure otherwise and constitute a valuable addition to the literature. The results of the N_2/H_2 measurements are reported in a paper recently submitted to JASA. The measurements in N_2/CH_4 mixtures were reported at the December meeting of the Acoustical Society of America. In the case of N_2/CH_4 mixtures, the effect of spark-induced-reaction contaminants on the relaxation time in the gas mixture is evident.

From the results obtained to date, it is concluded that the SACER phenomena needs to be considered in many non-equilibrium situations. It is likely involved in producing instabilities in excited gases in a variety of circumstances.

Optoacoustic Studies of Liquids

Optoacoustic studies of SF_6 excited with a CO_2 laser were undertaken to investigate the effect of strong optical absorption. This work, published as Ref. 9, show a very interesting effect in addtion to including the first successful technique for predicting the optoacoustic signal in the presence of strong optical absorption. Specifically, it was found that as the gas pressure decreased, the acoustic overpressure steadily decreased until a pressure of about 200 mtorr was reached. Near 200 mtorr, the acoustic overpressure went through zero and became negative. That is, absorption of optical energy resulted in net cooling of the gas!

At first glance (and even second) cooling of the gas as a result of energy deposition appears to violate several if not all the laws of thermodynamics. Microscopically, this effect can be understood as follows. When radiation is absorbed by an SF_6 molecule, some molecules are excited from the ground state to the $\nu_3 = 1$ state. The $\nu_3 = 1$ state is closely coupled to the $\nu_4 + \nu_6 = 1$ state which is slightly higher in energy. At high pressure, during a few collisions, molecules in the $\nu_3 = 1$ state transfer to the $\nu_4 + \nu_6 = 1$ state with the additional energy required coming from translation (the gas first cools). As energy is transferred down the manifold of vibrational states in subsequent collisions, the gas heats. At low pressures, however, collisions at the container walls become more frequent. At about 200 mtorr, the $\nu_3 = 1$ to $\nu_4 + \nu_6 = 1$ transfer is more probable than a wall collision (the gas cools), but transfer of $\nu_4 + \nu_6 = 1$ to the walls is more probable than transfer down through the manifold of vibrational states (there is no subsequent heating). The excess energy is carried off by the cell walls (heat sink). A set of energy transfer rate constants which predict the high pressure optoacoustic signal in SF_6 reliably predicts the transition from heating to cooling.

Although these results in SF_6 left us excited as well as confused, we could not imagine an encore performance. All work on the gas phase was discontinued in 1984. Fortified with the excellent agreement between theory and experiment in the gas phase, measurements in liquids began in earnest in 1984.

There are numerous publications reporting optoacoustic measurements in fluids. By and large, these papers are concerned with the amplitude of the optoacoustic signal in different fluids or with differing laser beam geometries. Our goal is to use optoacoustics as a tool to study the molecular processes which give rise to the acoustic signal then optimize the acoustic amplitude with this microscopic insight.

The basic problem associated with quantifying molecular dynamics in the liquid state is that such processes are extremely fast and there is no experimental parameter (such as pressure) to slow these processes down. The speed of the process to be studied placed severe (perhaps insurmountable) constraints on our experimental system. The best time resolution published as of 1984 was that achieved by Sullivan and Tam [10]. Our initial strategy was to reproduce the results of Sullivan and Tam and then improve temporal resolution.

An optoacoustic signal in propanol was first observed in our laboratory in the fall of 1985. By the fall of 1986, the basic results of Ref. 10 had been reproduced in propanol with a red dye and other alcohols. In addition, using a broadband hydrophone, the absolute magnitude of the optoacoustic signal was found to agree well with theoretical predictions. The decrease in optoacoustic amplitude with distance from the excitation zone was also found to agree with theory. Measurements were also made of optoacoustic amplitude for different optical penetration depths and input energies. Each observation agreed well with theory. This series of measurements was presented at the Fall, 1986 meeting of the Acoustical

Society of America. We have yet to decide if the results reported warrant a separate publication.

At the end of 1986, we were comfortable with the technique to measure very rapid conversion of electromagnetic radiation into an acoustic pulse. The next goal is to improve temporal resolution so that microscopic molecular transfer times can be determined. This might require identification of a fluid which reacts more slowly than propanol or water.

Summary

This six year study of Optical Sound Generation and Amplification has yielded some exciting and unexpected results. These include the first observation of sound amplification following electrical excitation of a gas, cooling of SF_6 following addition of energy, and a thorough verification of the theory of the spectrophone in the gas and fluid states.

Much work remains to be done. Our attention will next focus on other amplification mechanisms and controlled optoacoustic measurements in the regime where the fluid begins to vaporize. These measurements should shed new light on the transport of energy and molecules in the gas and fluid states. But our work is far from comprehensive when compared to ongoing research in other countries. The acoustic analog to four-wave mixing and other non-linear optical phenomena represents a large area to be explored by the acoustics community. It is our hope that an increased level of effort in this laboratory or others in this country will soon be possible, opening new frontiers for the old science of physical acoustics.

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